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Physics Department, W.S.C.

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A Study of the Far Infrared Properties of Crystals

by

J. H. ROHRBAUGH

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A STUDY OF THE FAR INFRARED PROPERTIES OF CRYSTALS

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Scientific Report #16

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Submitted by

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Abstract

A modification of Simon's method is described for determining the complex index of refraction from reflection measurements. Polarization studies involving a model scaled from the infrared to the millimeter wave are reported wherein the diffraction resulting from various strips is obtained experimentally and theoretically. The eigenvalues for a subdivision of 24 in wave vector space are also given for NaCl.

The Far Infrared Spectrometer

Considerable thought has been given to the problems involved in making accurate determinations of the complex index of refraction of crystals in the wavelength regions of interest to us. In the usual method, which necessitates measurement of both the reflection and the transmission coefficients, two samples are required, a thick crystal for the reflection measurements and a thin crystal for the transmission measurements. However, in regions of strong absorption, it becomes necessary to make the crystal exceedingly thin in order to permit any transmission at all. Most previous workers have resorted to evaporating films on substrates and then making their transmission measurements on the composite. This, of course, introduces problems in connection with the infrared properties of the substrate. Also, and perhaps more serious, is the question as to how the properties of the crystal in the form of evaporated film relate to those for the crystal in bulk and, therefore, whether the measurements have meaning at all.

In order to avoid these difficulties and the concomital uncertainty in interpretation, it has been decided to employ a modification of a method described by Simon[1], which permits determination of the complex index of refraction of the crystal from reflection measurements alone on a single thick crystal. Simon's method involves measurement of the reflection coefficient for two different angles of incidence. From these two reflection measurements Simon was able to determine both the refractive index and extinction coefficient of the crystal, thereby obviating

the need for any transmission measurement.

We have devised a modification of Simon's procedure which eliminates the need for any extensive reconstruction of the exit system of our spectrometer as would be required if we had to allow for rotation of the detector and associated exit optics in connection with the variation of the angular orientation of the crystal. Instead, we will use a crystal mounting in the form of a 90° corner reflector, which has the property of returning radiation incident at any angle in a given plane in the same direction from whence it came, but displaced laterally. By placing the crystal alternately in the two arms of the corner reflector with a plane mirror in the other arm each time, we will be able to obtain measurements at any two complementary angles without the need for rotating any part of our system. The measurement procedure to be followed will consist of point-by-point measurements over the wavelength range of a given grating, at each such point the signal level to be measured for the crystal alternately in the two arms of the corner reflector and, finally, a signal level measurement with plane mirrors in both arms of the corner reflector, this information sufficing to determine the reflection coefficint for each of the desired complementary angles of incidence, say, for example, 20° and 70°. From these two values of the reflection coefficient the complex index of refraction can then be determined for that particular wavelength, following Simon. The corner reflector mounting is now being built and will be installed in the system shortly. The crystal measurements will begin immediately thereafter.

Simon's method requires, in addition, a knowledge of the

apparatus polarization, that is, the polarization introduced into an initially unpolarized beam due to its passage through the system. From the previous work with echelette gratings in this laboratory[2,3], we know that our gratings will act as polarizing agents. These earlier experiments did not, however, include a study of the variation of the polarizing factor as function of angle of incidence or as a function of the number of grating rulings illuminated. In order to obtain this necessary information, it was decided to reactivate the millimeter wave spectrometer temporarily, and to perform these experiments. The apparatus used previously was modified and partially-rebuilt so as to give us greater accuracy and flexibility for the purposes of the present experiment. We had found in our previous work that attempting to mask rulings of a grating so as to delineate a given number of rulings led to error, since the masking is difficult to accomplish effectively. Consequently, a different procedure is being followed in the present work. We employ a frame upon which metal strips can be placed, and by making measurements with one strip, two strips, three strips, etc., in turn we can study the variation of the polarizing factor with the number of strips illuminated without introducing error due to undesired reflection from inadequately-masked strips. The apparatus provides, of course for variation of angle of incidence also. These measurements are now in progress. Though subsidiary to our main task, it

would seem that these experiments may be of interest to other people in the field, and we shall therefore include the results of this work in this and in the next report.

Methods of Theoretical Physics

For the case of the single strip "grating" work was done here which indicates that the diffraction of the strip is a function of the direction of polarization of the incident radiation on the diffracting element. Theoretically the case of the single strip is examined by Morse and Feshbach[4] who present the exact solutions for radiation incident on a plane infinite thin strip. These solutions are quite different for the situation where the incident radiation is polarized perpendicular to the strip as compared to the case where the polarization is parallel to the strip.

With the aid of a 704 Computer, we have been able to construct graphs of these exact solutions for a strip whose width is representative of the grating constants of the gratings used in our infrared spectrometer and for an angle of incidence which is intermediate in value to those used in our infrared experiments.

Very good experimental verification was obtained for the situation where the incident radiation is polarized in a direction which is parallel to the long side of the strip. The other situation, radiation polarized perpendicular to the strip does not agree quite so closely with the theoretical curve. This is the polarization, however, which yields large

anomalies when considered with classical diffraction theory. The former results, however, are extremely good and definitely indicate that our apparatus is functioning properly. Theoretical and experimental curves are shown in figures 1 and 2 (pp. 32 and 33).

Data has also been taken for both polarizations for gratings of from two to five strips and is now in the process of being graphed and analyzed. The experimental results for these built up gratings are of utmost significance since they are extremely difficult if not impossible to obtain in an exact form theoretically. The theoretical superposition of the patterns of individual strips without taking into account the interaction of the surfaces usually not considered in such a simple superposition cannot give exact results. The analysis of the experimental superposition of the diffraction patterns of a number of strips will therefore yield valuable information in helping us to determine the polarizing properties of an echelette grating and will in turn permit more accurate determinations of the complex indices of refraction of crystals using a spectrometer which has such a grating as its diffracting element.

The Characteristic Lattice Frequencies of NaCl

As described in the previous report (Scientific Report #15), we have set up a program for calculating the Coulomb terms and the consequent eigenfrequencies and eigenvectors for a Kellerman model NaCl lattice. The allowed region in wave-vector space was subdivided into 24 parts for each co-ordinate axis.

This gives rise to 82,944 frequencies. Due to the symmetry of the facecentered cubic structure calculations are needed for only 423 choices of the wave vector $\vec{\sigma} = \frac{1}{2r_o}(q_x, q_y, q_z) = \frac{1}{2r_o}\ \frac{1}{24}(p_x, p_y, p_z)$ with p_x, p_y, p_z positive integers such that $p_x + p_y + p_z \leqslant 36$ and $24 \geqslant p_x \geqslant p_y \geqslant p_z \geqslant 0$. The table below gives the six circular frequencies $\omega = 2\pi\nu$ scaled by 10^{-10} for each of the 423 vectors denoted by the value of p_x, p_y, p_z to the left. This choice of wave-vector density compares to a subdivision of $\vec{\sigma}$ into 10 parts by Kellerman giving 48 vectors and to the previous work here where σ was divided into 12 parts giving 74 vectors. The computation time was considerably reduced with the use of an IBM 704 rather than UNIVAC which had been used before. For a few cases the matrix elements were slightly altered in order to avoid degeneracy of the eigenvalues for which the program could not properly obtain the eigenvectors. The effect on the eigenvalues due to these changes is very small. The eigenvectors which were also obtained are not included because they are not of immediate interest and also there are too many of them (some 15,000 numbers). Frequency distribution functions and specific heat calculations based on the frequencies given in the table below will follow in subsequent reports.

24 12 0	3385	3385	2872	2723	2723	2312
24 10 2	3541	3302	2858	2836	2627	2188
24 10 0	3561	3248	2891	2847	2550	2269
24 8 4	3659	3306	2893	2814	2637	1946
24 8 2	3727	3200	2920	2892	2466	2058
24 8 0	3751	3154	2933	2929	2341	2160
24 6 6	3702	3318	2910	2787	2653	1833
24 6 4	3822	3213	2952	2862	2472	1844
24 6 2	3900	3134	2976	2932	2272	1923
24 6 0	3926	3092	2984	2973	2128	2025
24 4 4	3953	3143	2990	2916	2271	1799
24 4 2	4037	3088	3012	2966	2080	1821
24 4 0	4066	3052	3019	3001	1944	1897
24 2 2	4125	3053	3033	2995	1918	1778
24 2 0	4155	3039	3029	3017	1817	1806
24 0 0	4186	3046	3022	3022	1772	1772
23 11 1	3485	3319	2875	2785	2646	2274
23 9 3	3632	3283	2886	2825	2612	2058

23	9	1	3667	3206	2924	2859	2477	2184
23	7	5	3717	3293	2916	2792	2632	1859
23	7	3	3802	3196	2945	2869	2460	1929
23	7	1	3849	3129	2972	2919	2277	2049
23	5	5	3851	3204	2957	2855	2465	1810
23	5	3	3952	3135	2985	2917	2267	1827
23	5	1	4006	3080	3005	2965	2081	1912
23	3	3	4062	3089	3010	2960	2074	1782
23	3	1	4119	3050	3025	2996	1916	1810
23	1	1	4179	3040	3026	3014	1806	1770
22	14	0	3541	3302	2858	2836	2627	2188
22	12	2	3514	3337	2844	2828	2611	2226
22	12	0	3448	3371	2868	2722	2710	2263
22	10	4	3634	3311	2908	2790	2593	2054
22	10	2	3621	3261	2901	2800	2575	2170
22	10	0	3625	3238	2921	2783	2534	2241
22	8	6	3709	3316	2948	2723	2641	1875
22	8	4	3748	3238	2930	2799	2580	1932

22	8	2	3791	3175	2950	2842	2438	2044
22	8	0	3807	3147	2970	2853	2324	2140
22	6	6	3792	3237	2945	2786	2592	1825
22	6	4	3886	3171	2955	2853	2440	1832
22	6	2	3951	3116	2981	2903	2250	1910
22	6	0	3974	3086	3004	2918	2111	2008
22	4	4	4002	3122	2977	2911	2250	1783
22	4	2	4079	3079	2998	2954	2062	1806
22	4	0	4105	3047	3024	2968	1927	1881
22	2	2	4157	3051	3003	2991	1901	1763
22	2	0	4190	3031	3025	3002	1802	1790
22	0	0	4218	3028	3018	3017	1757	1756
21	13	1	3501	3373	2841	2806	2652	2174
21	11	3	3658	3268	2922	2797	2521	2138
21	11	1	3613	3282	2900	2741	2587	2214
21	9	5	3778	3232	2982	2742	2543	1937
21	9	3	3777	3195	2942	2784	2516	2033
21	9	1	3786	3180	2948	2771	2433	2146

21	7	7	3821	3229	3000	2701	2578	1836
21	7	5	3869	3169	2966	2782	2532	1837
21	7	3	3922	3134	2957	2830	2401	1903
21	7	1	3952	3110	2981	2844	2238	2017
21	5	5	3970	3127	2954	2848	2404	1785
21	5	3	4050	3096	2965	2897	2223	1800
21	5	1	4092	3066	2999	2912	2045	1882
21	3	3	4143	3068	2970	2952	2038	1753
21	3	1	4192	3041	3005	2964	1882	1779
21	1	1	4245	3024	3006	2995	1774	1738
20	16	0	3659	3306	2893	2814	2637	1946
20	14	2	3634	3311	2908	2790	2593	2054
20	14	0	3486	3443	2818	2802	2699	2065
20	12	4	3773	3228	2992	2748	2414	2087
20	12	2	3660	3289	2914	2768	2529	2141
20	12	0	3611	3330	2880	2671	2667	2163
20	10	6	3888	3232	3008	2670	2415	1942
20	10	4	3826	3185	2989	2745	2451	2020

20	10	2	3800	3198	2936	2730	2475	2118
20	10	0	3792	3210	2926	2686	2485	2171
20	8	8	3928	3248	3004	2615	2462	1857
20	8	6	3917	3157	3017	2705	2491	1847
20	8	4	3935	3114	3974	2767	2459	1901
20	8	2	3954	3122	2955	2766	2362	2001
20	8	0	3962	3127	2957	2758	2273	2086
20	6	6	3983	3089	2989	2774	2470	1790
20	6	4	4048	3078	2949	2831	2355	1795
20	6	2	4093	3074	2964	2843	2189	1867
20	6	0	4107	3070	2981	2842	2061	1959
20	4	4	4140	3061	2933	2901	2184	1745
20	4	2	4200	3047	2967	2912	2007	1762
20	4	0	4220	3033	2996	2911	1879	1834
20	2	2	4267	3030	2979	2952	1851	1717
20	2	0	4291	3012	3004	2955	1753	1743
20	0	0	4315	3005	3005	2970	1709	1709
19	15	1	3589	3395	2874	2771	2678	1956

19 13 3	3762	3255	2984	2730	2462	2043
19 13 1	3647	3351	2884	2729	2605	2065
19 11 5	3924	3219	2990	2682	2331	2003
19 11 3	3854	3193	2975	2712	2406	2075
19 11 1	3816	3234	2909	2655	2509	2125
19 9 7	4014	3252	2949	2621	2372	1861
19 9 5	3993	3128	2994	2701	2398	1899
19 9 3	3988	3108	2961	2712	2392	1984
19 9 1	3986	3139	2924	2668	2356	2094
19 7 7	4042	3130	2976	2691	2423	1800
19 7 5	4083	3023	2991	2760	2397	1795
19 7 3	4117	3053	2943	2773	2300	1853
19 7 1	4134	3080	2941	2759	2164	1957
19 5 5	4164	3018	2927	2835	2298	1737
19 5 3	4222	3028	2927	2855	2139	1746
19 5 1	4251	3038	2955	2844	1976	1822
19 3 3	4296	3018	2937	2901	1965	1696
19 3 1	4334	3013	2971	2899	1817	1718

19	1	1	4376	3000	2990	2919	1711	1677
18	18	0	3702	3318	2910	2787	2653	1833
18	16	2	3709	3316	2948	2723	2641	1875
18	16	0	3555	3462	2843	2785	2708	1884
18	14	4	3888	3232	3008	2670	2415	1942
18	14	2	3728	3321	2950	2700	2559	1969
18	14	0	3643	3404	2854	2740	2634	1975
18	12	6	4049	3304	2901	2608	2222	1971
18	12	4	3939	3217	2993	2656	2323	2017
18	12	2	3865	3235	2942	2658	2452	2051
18	12	0	3836	3271	2883	2593	2578	2063
18	10	8	4166	3283	2848	2527	2292	1861
18	10	6	4079	3250	2903	2625	2279	1906
18	10	4	4048	3124	2978	2659	2320	1968
18	10	2	4027	3141	2926	2630	2366	2046
18	10	0	4019	3169	2894	2583	2402	2082
18	8	8	4129	3273	2858	2602	2310	1818
18	8	6	4148	3126	2911	2679	2340	1802

18	8	4	4166	3016	2964	2705	2321	1845
18	8	2	4175	3070	2913	2680	2253	1935
18	8	0	4178	3097	2898	2662	2189	2006
18	6	6	4215	2993	2927	2761	2323	1740
18	6	4	4267	2974	2914	2794	2233	1735
18	6	2	4296	3024	2906	2773	2091	1799
18	6	0	4305	3046	2907	2762	1978	1882
18	4	4	4340	2977	2882	2862	2083	1678
18	4	2	4382	2998	2923	2840	1919	1691
18	4	0	4397	3012	2932	2833	1798	1757
18	2	2	4435	2987	2960	2857	1769	1643
18	2	0	4452	2992	2969	2859	1675	1665
18	0	0	4471	2986	2985	2865	1632	1631
17	17	1	3624	3407	2893	2732	2707	1855
17	15	3	3830	3273	3008	2641	2522	1891
17	15	1	3685	3395	2897	2712	2620	1906
17	13	5	4039	3293	2927	2588	2267	1945
17	13	3	3934	3233	2992	2610	2402	1971

17	13	1	3869	3282	2897	2622	2528	1985
17	11	7	4180	3373	2783	2542	2154	1910
17	11	5	4124	3232	2910	2587	2219	1953
17	11	3	4084	3138	2964	2590	2304	2005
17	11	1	4059	3179	2883	2546	2414	2036
17	9	9	4230	3404	2725	2512	2176	1842
17	9	7	4225	3273	2789	2592	2231	1818
17	9	5	4231	3109	2897	2626	2256	1845
17	9	3	4231	3045	2926	2614	2260	1916
17	9	1	4227	3101	2865	2577	2246	1992
17	7	7	4283	3121	2811	2676	2267	1748
17	7	5	4328	2975	2887	2718	2245	1733
17	7	3	4353	2987	2882	2701	2168	1780
17	7	1	4364	3045	2852	2675	2058	1871
17	5	5	4399	2906	2859	2816	2158	1667
17	5	3	4444	2957	2864	2789	2021	1668
17	5	1	4464	3007	2868	2762	1875	1735
17	3	3	4501	2950	2916	2797	1860	1612

17 3 1	4529	2982	2925	2789	1721	1629
17 1 1	4559	2971	2967	2792	1617	1585
16 16 4	3928	3248	3004	2615	2462	1857
16 16 2	3758	3337	2966	2627	2619	1877
16 16 0	3657	3432	2848	2763	2625	1887
16 14 6	4138	3364	2828	2533	2227	1879
16 14 4	4009	3269	2976	2550	2363	1907
16 14 2	3915	3268	2957	2575	2500	1925
16 14 0	3876	3309	2866	2645	2538	1931
16 12 8	4281	3464	2657	2472	2056	1904
16 12 6	4201	3350	2808	2501	2136	1930
16 12 4	4145	3199	2947	2527	2240	1961
16 12 2	4104	3172	2923	2524	2368	1980
16 12 0	4085	3203	2850	2483	2481	1988
16 10 10	4333	3501	2596	2430	2046	1868
16 10 8	4305	3410	2659	2504	2110	1842
16 10 6	4299	3247	2792	2539	2154	1856
16 10 4	4292	3082	2919	2542	2194	1906

16	10	2	4280	3087	2873	2518	2246	1963
16	10	0	4274	3121	2822	2489	2285	1988
16	8	8	4352	3270	2679	2590	2166	1771
16	8	6	4395	3097	2781	2627	2184	1742
16	8	4	4418	2958	2892	2620	2167	1774
16	8	2	4426	3024	2824	2594	2118	1848
16	8	0	4426	3060	2799	2576	2072	1906
16	6	6	4468	2945	2783	2741	2160	1667
16	6	4	4514	2891	2838	2737	2082	1655
16	6	2	4536	2982	2800	2697	1963	1707
16	6	0	4542	3015	2794	2679	1865	1781
16	4	4	4576	288[illegible]	2861	2757	1947	1590
16	4	2	4608	2954	2862	2722	1800	1594
16	4	0	4618	2985	2860	2711	1689	1654
16	2	2	4648	2941	2935	2713	1658	1541
16	2	0	4660	2966	2933	2708	1568	1560
16	0	0	4673	2960	2960	2707	1525	1525
15	15	5	4080	3325	2907	2521	2307	1875

15	15	3	3966	3255	2996	2529	2460	1897
15	15	1	3892	3302	2899	2614	2533	1909
15	13	7	4273	3445	2697	2452	2085	1894
15	13	5	4202	3299	2873	2464	2192	1917
15	13	3	4147	3177	2963	2473	2326	1934
15	13	1	4112	3204	2868	2490	2444	1943
15	11	9	4377	3515	2538	2427	2004	1868
15	11	7	4358	3379	2681	2455	2053	1873
15	11	5	4346	3197	2845	2464	2110	1903
15	11	3	4330	3076	2917	2461	2195	1938
15	11	1	4315	3121	2817	2441	2294	1957
15	9	9	4412	3410	2551	2502	2059	1801
15	9	7	4454	3240	2669	2529	2091	1765
15	9	5	4480	3053	2822	2527	2106	1779
15	9	3	4487	2990	2850	2515	2111	1835
15	9	1	4486	3056	2762	2494	2109	1893
15	7	7	4534	3067	2660	2659	2100	1681
15	7	5	4584	2901	2804	2648	2073	1654

15	7	3	4609	2937	2771	2627	2010	1689
15	7	1	4616	3008	2724	2599	1922	1765
15	5	5	4654	2794	2789	2765	1993	1577
15	5	3	4692	2906	2782	2680	1874	1568
15	5	1	4706	2973	2777	2638	1746	1624
15	3	3	4738	2890	2884	2642	1726	1504
15	3	1	4758	2951	2878	2621	1597	1514
15	1	1	4781	2940	2939	2612	1497	1467
14	14	8	4333	3501	2596	2430	2045	1868
14	14	6	4244	3386	2781	2438	2142	1895
14	14	4	4182	3231	2938	2444	2270	1912
14	14	2	4133	3187	2924	2450	2417	1925
14	14	0	4115	3216	2839	2509	2450	1927
14	12	10	4438	3578	2436	2375	1933	1882
14	12	8	4402	3486	2574	2392	1977	1888
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14 10 8	4496	3375	2564	2437	1998	1799
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5	1	1	5820	2803	2792	1063	597	581
4	4	4	5769	2720	2719	1390	818	818
4	4	2	5813	2782	2711	1195	739	698
4	4	0	5822	2816	2709	1117	713	653
4	2	2	5864	2780	2753	986	598	566
4	2	0	5873	2804	2760	908	538	508
4	0	0	5880	2800	2800	834	446	446

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3 3 1	5892	2793	2741	873	544	503
3 1 1	5924	2789	2779	679	393	379
2 2 2	5931	2769	2768	697	419	416
2 2 0	5945	2793	2765	572	356	321
2 0 0	5959	2789	2788	421	229	222
1 1 1	5972	2782	2780	343	219	206

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4. Morse and Feshbach Methods of Theoretical Physics Vol.II

Fig. 1

Diffraction From A Single Strip

Theoretical Curve

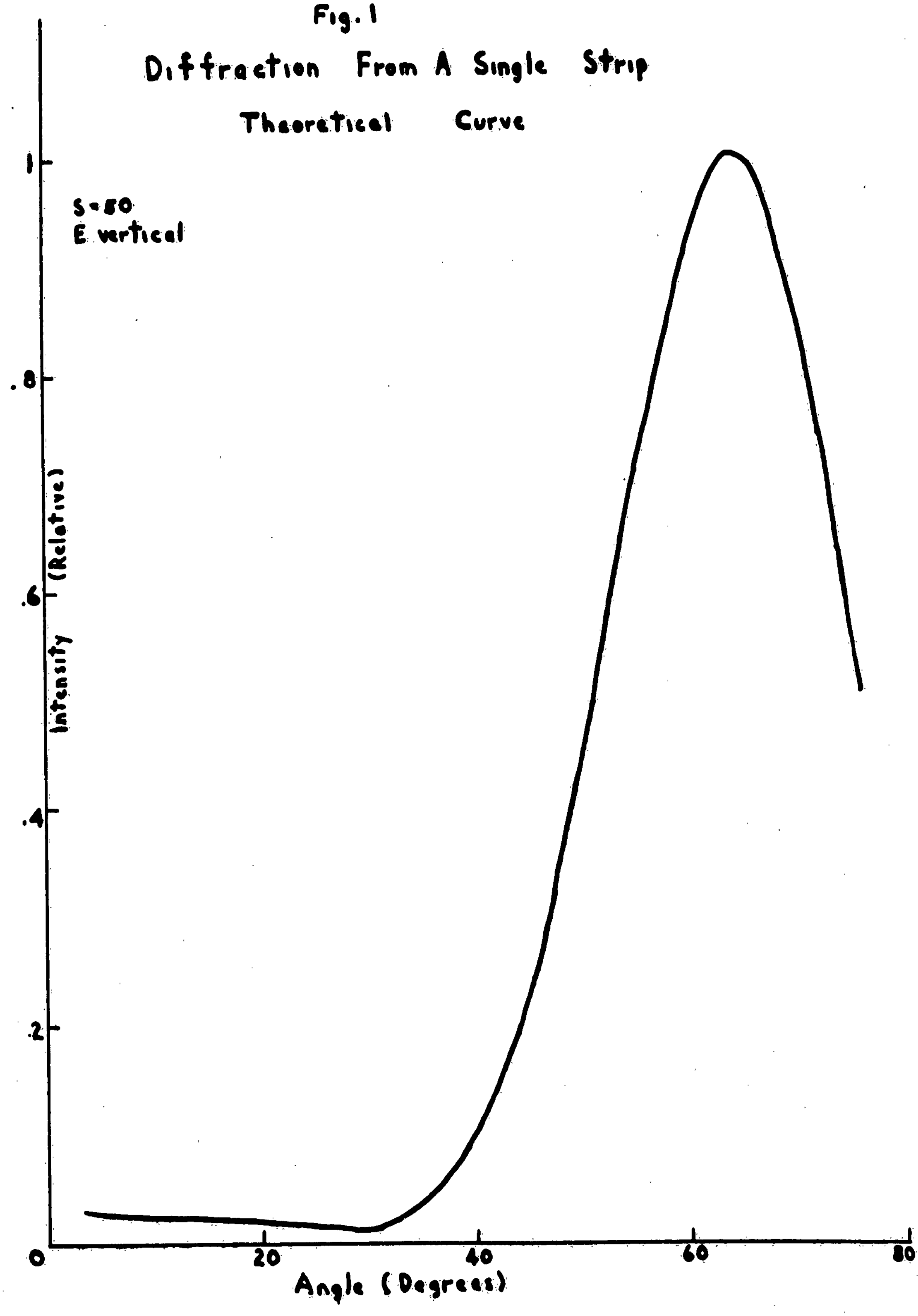
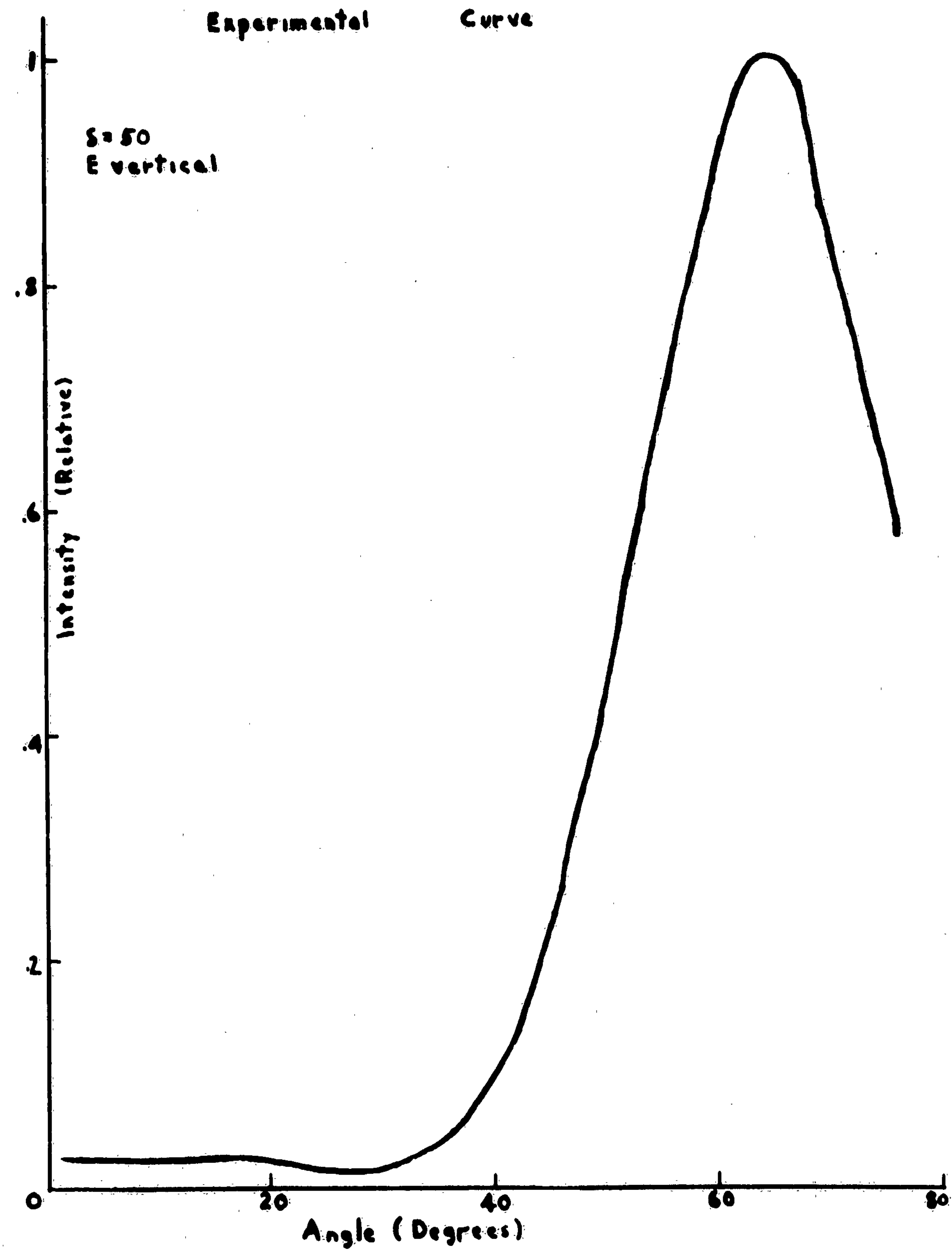


Fig. 2

Diffraction From A Single Strip



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